

# Role of Coulomb correlation on magnetic and transport properties of doped manganites: $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ and $\text{LaSr}_2\text{Mn}_2\text{O}_7$

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## Abstract

Results of LSDA and LSDA+U calculations of the electronic structure and magnetic configurations of the 50 % hole-doped pseudocubic perovskite  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and double layered  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  are presented. We demonstrate that the on-site Coulomb correlation (U) of Mn *d* electrons has a very different influence on the *(i)* band formations, *(ii)* magnetic ground states, *(iii)* interlayer exchange interactions, and *(iv)* anisotropy of the electrical transport in these two manganites. A possible reason why the LSDA failures in predicting observed magnetic and transport properties of the double layered compound - in contrast to the doped perovskite manganite - is considered on the basis of a p-d hybridization analysis.

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## 1 Introduction

The half-doped manganites,  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ , belong to the Ruddlesden-Popper family [1],  $(\text{R,A})_{n+1}\text{Mn}_n\text{O}_{3n+1}$ , which has been attracting intensive interest due to itinerant doped carriers that give rise to a variety of interesting physical phenomena, notably colossal magnetoresistance. One of the most striking properties of the double layered manganites,  $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ ,

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is the extremely large magnetoresistance which is much larger than that for prototypical perovskite systems: it was found in 40 % doped  $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  that the resistivity ratio reaches  $\sim 20,000$  % at 7 T above  $T_c$  [2], whereas for  $\text{La}_{0.6}\text{Sr}_{0.4}\text{MnO}_3$   $\rho(0)/\rho(H) \sim 200$  % at 15 T [3]. The main difference between 3D perovskite and 2D layered compounds arises from the change of dimensionality ( $n$ ) of the crystal structure connected with the number of  $\text{MnO}_6$  layers along the  $c$  direction ( $n=\infty$  for pseudocubic perovskite systems and  $n=2$  for double layered manganites), which in turn has a strong influence on their magnetic and transport properties [2, 4, 5, 6, 7]. Measurements and comparison of characteristic features (e.g., the strong anisotropy of electrical (magneto-) transport and magnetostriction, and the 2D character of magnetism caused by reduction of the exchange coupling between the Mn ions along the  $c$  direction in the double-layered manganites) suggested that both the anisotropic transfer interaction and the two dimensional spin correlation have a strong influence on the CMR properties in layered manganites [2].

The pseudocubic perovskites,  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ , have an insulating paramagnetic phase at high temperature and a metallic ferromagnetic phase at low temperature over a wide range of hole doping [3, 8]. The strong coupling between the magnetic ordering and the electrical conductivity demonstrates the strong relationship between the electrical resistivity and the spin alignment which has been explained by the double-exchange mechanism arising from the mixed valence of  $\text{Mn}^{3+}/\text{Mn}^{4+}$ . In comparison to these pseudocubic perovskites, the double layered structures ( $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ ) with their two-dimensional networks of  $\text{MnO}_6$  octahedra have a reduced exchange coupling between the Mn ions along the  $c$  direction. Indeed, in the double-layered case consisting of two perovskite blocks separated by an intervening insulating layer of (La,Sr)O ions along the  $c$  axis, the balance between antiferromagnetism and ferromagnetism is very sensitive to  $e_g$  band filling [9]. Bilayer  $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  demonstrates ferromagnetism in a doping range  $x < 0.39$ , a canted antiferromagnetic structure for a hole concentration  $0.39 < x < 0.48$ , and layered antiferromagnetic states for  $x > 0.48$  [10, 11].

An important issue in the theory of CMR oxides is the role of Coulomb correlation since an accurate treatment of correlation may affect significantly the balance between ferromagnetic and antiferromagnetic interactions and hence the magnetic ground state. According to our previous investigations [12], where we demonstrated that the on-site Coulomb correlation of Mn  $d$  electrons significantly modifies the electronic structure, magnetic ordering, interlayered exchange interactions and promotes strong electronic transport anisotropy in 50 % doped  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ , the experimentally observed magnetic ordering is reproduced within LSDA+U calculations only for  $U \geq 7$  eV. This is in contrast with the CMR perovskite 3D manganites ( $\text{LaMnO}_3$ ,  $(\text{La,Sr})\text{MnO}_3$  systems) where LSDA theory correctly predicts the magnetic ground state [13, 14].

Considering two 50 % doped manganites, the double layered  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  and pseudocubic perovskite  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ , we outline here the main similarities and differences in their characteristic properties: (*i*) Both manganites formally consist of  $\text{Mn}^{3.5+}$  ions surrounded by oxygen octahedra, and so in a

high spin configuration the majority  $t_{2g}$  states are filled and the electrically active orbitals are  $d_{x^2-y^2}$  and  $d_{3z^2-r^2}$ . (ii) The dynamical Jahn-Teller (JT) effect is absent for both systems,  $\Delta_{JT} = D(Mn - O_{apical})/D(Mn - O_{planar})$ , where  $D(Mn - O)$  is manganese oxygen distance, equals to 1.004 and 1.001 for  $La_{0.5}Sr_{0.5}MnO_3$  and  $LaSr_2Mn_2O_7$ , respectively – although it should be noted here that due to an intervening layer of (La.Sr)O ions along the  $c$  axis in the latter manganite, its  $MnO_6$  octahedra are slightly distorted in terms of different distances between Mn and apical oxygens. Hence, in contrast to the 3D perovskite manganites which have two types of oxygen atoms (one apical and one planar), there are three oxygen types in the double layered manganites – two apical and one planar. (iii) The resistivity of  $La_{0.5}Sr_{0.5}MnO_3$  is of the order of  $10^{-3}\Omega cm$  with a flat temperature dependence [8], whereas for  $LaSr_2Mn_2O_7$  the  $ab$ -plane resistivity is  $10^0-10^{-1}\Omega cm$  [15] and the resistivity perpendicular to the layer is  $\sim 10^2$  times larger; in addition, according to previous theoretical works [14, 12, 16] both are half-metals. (iv) They have different magnetic orderings:  $La_{0.5}Sr_{0.5}MnO_3$  is a ferromagnet (FM), while  $LaSr_2Mn_2O_7$  is an A-type antiferromagnet (AFM) (magnetic moments lie in the  $ab$  plane and couple ferromagnetically within the single  $MnO_2$  layer, but show AFM order between layers). (v) As mentioned above, the different influence of the on-site Coulomb correlation on the magnetic ordering in these manganites has been established in the previous calculations.

In this paper, on the basis of comprehensive analyses of the crystal structures, electronic structures and magnetic configurations of the double layered  $LaSr_2Mn_2O_7$  and pseudocubic perovskite  $La_{0.5}Sr_{0.5}MnO_3$  compounds, we establish the mechanisms governing the band formation in these manganites and the different role of the on-site Coulomb correlation in its relation with the electronic, magnetic and transport properties. For both compounds, we consider the total energy differences between FM and AFM spin configurations for the Coulomb correlation parameter,  $U$ , equal to 0 and 7 eV; then, for the ground states of these manganites, we compare the exchange interaction parameters and the most important band characteristics (such as the population of Mn-d states near the Fermi level, Mn-d sub-band widths and the p-d hybridization) and give a possible reason why the LSDA scheme ( $U=0$ ) fails to predict the magnetic ground state as well as the electrical transport properties for the double layered manganite, in contrast to its success for the case of the pseudocubic perovskite manganite.

## 2 Methodology

The LSDA and LSDA+ $U$  calculations were performed in the frame-work of the linear-muffin-tin-orbital method in the atomic sphere approximation (LMTO-ASA) [17]. We used the von Barth-Hedin-Janak form [18] for the exchange-correlation potential. The crystal parameters were taken from Ref. [19, 20]. From the constrained LSDA supercell calculations [21, 22], we obtained values of the Coulomb and exchange parameters to be  $U=7.1$  eV and  $J=0.79$  eV for

$\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $U=7.2$  eV and  $J=0.78$  eV for  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ . These values are typical for the transition-metal oxides [23, 24, 25].

### 3 Results and Discussion

#### 3.1 Electronic structure, transport anisotropy and magnetism

As a first step, we performed the band structure calculations of the FM (all atoms in every layer and between layers are ordered ferromagnetically) and the A-type AFM (ferromagnetic layers stacked antiferromagnetically) phases of both  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  by the standard LSDA method. FM spin alignment is found to be more preferable from total energy calculations for both manganites (Table 1). The projected densities of states (DOS) for these FM cases look very similar (Fig. 1): For the majority-spin channel, Mn 3d states of both compounds form the bands between 2.2 eV below  $E_F$  and 2.5 eV above  $E_F$ . As seen from the figure, the  $e_g$  ( $x^2 - y^2$  and  $3z^2 - r^2$ ) bands are partially filled, cross  $E_F$ , and are rather broad compared to the  $t_{2g}$  ( $xy$  and degenerate  $xz, yz$ ) bands which are about 1.2 eV wide and lie 1 eV below  $E_F$ . The exchange interaction splits the Mn 3d states such that the  $t_{2g}$  and  $e_g$  minority-spin bands are located 2.6 eV higher in energy than the majority-spin states. We obtained a band gap in the minority-spin  $d$  bands of around 2.5 eV for both  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  – in keeping with the fact that doped perovskite manganites are found to be half-metallic [26, 27, 14]. The analogs in the main band characteristics near  $E_F$  in the calculated electronic spectra of the double layered and doped perovskite manganites allow us to suggest that the insulating character of minority-spin electrons for the double-layered manganite may also render the conduction band a half-metallic one (i.e., 100 % spin polarization for the conduction electrons). Note that  $E_F$  in our spin-polarized band structure calculations of the double-layered manganite was found to lie at the bottom of the  $t_{2g}$  minority spin conduction band (which is in agreement with the full-potential LMTO calculations for  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  [16]), and so the LSDA electronic band structure just misses to being half-metallic.

The crucial role of Coulomb correlation in the stability of the experimentally observed magnetic ground state of  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  was established in Ref. [12]. We investigated the electronic structure similarities of  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  and  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  for both FM and A-type AFM spin alignments in the framework of the LSDA+U method with the calculated  $U$  value of 7 eV. According to the total energy differences in Table 1, the ground state of the perovskite manganite remains ferromagnetic, while the observed A-type AFM ordering in double layered manganite is found to be energetically more favorable with  $U=7$  eV. (A possible reason for the LSDA failure for the case of the double layered manganite in contrast to the perovskite will be discussed later). A comparison of the projected DOS's for the LSDA+U ground states of  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  (solid line) and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  (dashed line) is shown in Fig. 2. Only majority

spin  $e_g$  states contribute to the DOS at  $E_F$ . We find the magnetic moment of the primitive unit cell to be equal to  $7.00 \mu_B$ . Thus, LSDA+U calculations result in a truly half-metallic state for the double layered manganite, and so electron conduction is possible only within the majority spin sublattice.

One of the main differences between the doped pseudocubic perovskite and double layered manganites which is clearly seen from Fig. 2, is the different behavior of the electrically active states when on-site Coulomb correlation is taken into account. The DOSs for the  $\text{LaSr}_2\text{Mn}_2\text{O}_7$   $e_g$  states demonstrate (Fig. 2) the significant difference between  $3z^2 - r^2$  and  $x^2 - y^2$  orbitals in the electron population near  $E_F$ , while for  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  the band widths and the position of the band centers of these two  $e_g$  orbitals as well as their contributions to the DOS at  $E_F$  are very close (Fig. 2).

In Table 1 we presented the ratio between the  $x^2 - y^2$  and  $3z^2 - r^2$  contributions to the DOS at  $E_F$  (denoted as  $N_{x^2-y^2}/N_{3z^2-r^2}$ ) calculated in both the LSDA and LSDA+U approaches. The ratio demonstrates the presence of anisotropy in the population of the two  $e_g$  states at  $E_F$ ; and since the situation does not change qualitatively in the 0.3 eV energy window just above  $E_F$  (Fig. 2), we draw the following conclusions in the picture of charge transport: For  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ , we have 3D conduction; the use of  $U=7$  eV does not result in a large splitting of the  $e_g$  states and the number of electrons that contribute to electrical transport remain almost insensitive to the influence of  $U$ . This is contrary to that in  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  where for  $U=7$  eV the electron conduction (hopping) along the  $c$  axis ( $N_{3z^2-r^2}$ ) becomes very small and thus, on-site correlation treated within LSDA+U promotes 2D type (in-plane) electronic behavior in the double layered system — as observed experimentally. Thus, the comparison of the LSDA and LSDA+U band structures for both manganites leads us to the following conclusion: the exchange splitting with almost unoccupied minority spin Mn-3d states and the ligand field splitting of the  $t_{2g}$  and  $e_g$  states are the main factors governing band formation in both  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ . However, further splitting of the  $e_g$  states with increase of  $U$  is an essential feature of the layered manganites, but is absent in  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ .

The total energy differences for the two manganites for  $U=0$  and 7 eV shown in Table 1 (where the total energy of the FM spin ordering for each  $U$  value is taken to be zero) demonstrate that for the double layered compound the on-site Mn-d electron Coulomb correlations modify the magnetic ordering from FM to A-type AFM — as experimentally observed, while the FM ground state of the pseudocubic perovskite remains energetically more preferred as  $U$  is increased. To determine the character of the exchange coupling in these manganites, we calculated the effective exchange interaction parameters (for computational details see [28]). In Table 1, the values of the d-d effective exchange parameters,  $J_{dd}$ , between nearest Mn neighbours which belong to the different layers of one bilayer are shown for both  $U$  values. As seen from the Table, they follow the magnetic ordering behavior (obtained by the total energy differences between FM and AFM configurations) for both compounds. (For more details of the change of the exchange interaction parameters and the FM-AFM total energy differences in  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  with the increase of  $U$ , see Ref. [12].)

We interpreted the calculated exchange coupling within the superexchange (SEX) and double exchange (DEX) models by calculating the  $e_g$ -sub-band widths of the manganite ground states. As can be seen from Table 1, where we present the ratio of the  $x^2 - y^2$  and  $3z^2 - r^2$  band widths (denoted as  $W_{x^2-y^2}/W_{3z^2-r^2}$ ), for  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  the  $e_g$ -band widths stay exactly equal with increase of  $U$ , and provide positive DEX interactions. We note here, that the increase of the ferromagnetic contributions to the exchange interaction energy (c.f., Table 1) may be due to the increase of the  $e_g$ -band widths from 3.4 eV for  $U=0$  to 3.8 eV for  $U=7$  eV and the stronger O-p hybridization in the Mn-d states – as will be illustrated below. In  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ , a sharp increase of the  $W_{x^2-y^2}/W_{3z^2-r^2}$  ratio coming from the narrowing of the  $3z^2 - r^2$  band width (Table 1 and Fig. 2) results in the strong splitting of these two  $e_g$ -sub-bands and so promotes the negative (AFM) contribution to the exchange interaction. Thus, Coulomb correlations have a very different influence on pseudocubic and double layered manganites: for the first we obtain an increase of the DEX contributions, while for  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ ,  $U$  suppresses the DEX contributions to the interlayer exchange interaction energy and produces the change in magnetic ordering from FM to AFM.

### 3.2 Significance of p-d hybridization

The degree of O-p hybridization with Mn-d states plays an important role in the band structure and on the exchange mechanism [13, 29]. We compared below the p-d hybridization of  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  for  $U=0$  and 7 eV by calculating the O-p contributions to the DOS at the energy windows of corresponding Mn-d states. And the comparison gives interesting differences between double layered and perovskite manganites, supporting the results on magnetic and transport properties we have already discussed above.

According to the qualitative picture of the simple DEX model, an electrically active electron has a finite hopping probability ( $t$ ) between ferromagnetically ordered Mn ions, but this hopping vanishes in the case of antiferromagnetic spin alignment due to strong Hund coupling ( $t < J$ ). The character of the occupied  $e_g$  orbitals, which affects transport and magnetic properties, has been established above, but it is also important to investigate the role of oxygen hybridization in the  $e_g$  and  $t_{2g}$  orbitals as well [29].

At first, we shall consider a significance of oxygen p-hybridization in the electrically active  $e_g$  orbitals. For both manganite compounds, two  $e_g$  ( $3z^2 - r^2$  and  $x^2 - y^2$ ) and  $p_x$ ,  $p_y$ ,  $p_z$  oxygen states contribute to the DOS at  $E_F$ . In Table 1, we present the percentage of these contributions for unit cells consisting of two Mn atoms for both compounds. As expected, only  $p_z$  orbitals of apical oxygens ( $p_x$ ,  $p_y$  of planar oxygens) hybridize with “out-of-plane”  $3z^2 - r^2$  (“in-plane”  $x^2 - y^2$ ) d-orbitals. Comparing the lowest spin configurations within LSDA and LSDA+ $U$  (as obtained by total energy differences), we draw the following conclusions: for ferromagnetic  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ , we have significant contributions to the DOS at  $E_F$  from both apical and planar oxygens, and the on-site Coulomb correlation resulting in a redistribution of the conduction electrons between  $e_g$

and p-states, do not change the 3D character of the hopping matrix (Table 1). The influence of  $U$  on the population picture at  $E_F$  in  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  is very different. Established above sharp decrease of the  $N_{3z^2-r^2}$  for  $U=7$  eV occurs together with vanishing apical oxygen contributions at  $E_F$  (Table 1) resulting in suppression by  $U$  the  $c$  axis transport. This leads to the superexchange antiferromagnetic coupling along the  $c$  axis – as was obtained by calculating the interlayer exchange interaction parameters (Table 1). In contrast, a sharp increase of the  $p_x$  ( $p_y$ ) hybridization in the  $x^2 - y^2$  orbital (due to the charge redistribution) provides strong DEX ferromagnetic interactions in the  $ab$ -plane.

Investigation of the oxygen contributions to the DOS at  $E_F$  allowed us to draw conclusions on the transport and exchange properties in  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ . Overlapping of Mn- $t_{2g}$  and O-p orbitals in these manganites is an essential factor governing their band formation. In order to compare the p- and d-band overlaps for the perovskite and double layered compounds, we should, first of all, compare Mn-O distances for these systems. The distances between Mn and apical and planar oxygens in  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  are 1.941 and 1.934 Å, respectively; for the double layered manganite they are  $\sim 0.01$  Å smaller than for the perovskite compound: 1.917 (1.940) Å for Mn and apical oxygens and 1.927 Å for Mn and planar oxygen distances. Such a negligible difference in Mn-O distances serves to stress our results below.

In Fig. 3, we present the calculated angular electron density distribution in the energy window of corresponding majority spin  $t_{2g}$  states for both manganites. As seen from the figure, the hybridization between Mn-d  $t_{2g}$  and O-p states for  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  exists for both  $U=0$  and 7 eV, becoming stronger as  $U$  is increased (Fig. 3(a) and 3(b)). The same result was obtained for undoped  $\text{LaMnO}_3$  [30], in which, however, strong Jahn-Teller distortions of  $\text{MnO}_6$  octahedra lead to more than two times larger oxygen contributions to  $t_{2g}$  Mn states than that for  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ . In contrast to these perovskites, and despite the almost equal Mn-O distances in the doped manganites, LSDA band structure calculations for  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  gave O-p contributions to the DOS at the energy windows corresponding to  $t_{2g}$  states that are almost negligible (Fig. 3(c)). The p-d hybridization for the double layered manganite is correctly described only treated in LSDA+ $U$  (Fig. 3(d)): it becomes very strong between Mn and planar oxygens and one of the apical atoms, O1 (which lies between Mn-layers of one  $\text{MnO}_6$  bilayer unit), while it is suppressed for apical O2 atoms which belong to the (La,Sr) insulating layer as well. Thus, we believe that the absence p-d hybridization as an important ingredient in the band formation picture results in the failure of the LSDA scheme in predicting observed magnetic and transport properties of the double layered manganite.

The lowered dimensionality of the magnetic interactions in the double layered system, which was established under the influence of the on-site Coulomb correlation, is also seen from a comparison of two LSDA+ $U$  cases for  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  (Fig. 3(b) and 3(d)): in the 3D-compound, we have equal contributions to the DOS from all p-states ( $p_x$ ,  $p_y$  and  $p_z$ ) of planar oxygens, while for the double layered manganite only  $p_x(p_y)$  states of the planar oxygens give significant contributions to the DOS at the energy window correspond-

ing to Mn-d  $t_{2g}$  states. In addition, the absence of the hybridization between Mn-d states and apical O2-p states (this oxygen lies at the boundary of the perovskite-type bilayer) makes the whole hybridization picture obtained for the double layered  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  for  $U=7$  eV (Fig. 3(d)) to be in a very good agreement with the structural peculiarity of this compound - namely the existence of two perovskite-type layers separated by a rock-salt-type layer  $(\text{La,Sr})_2\text{O}_2$ .

## 4 Summary

In summary, the calculated electronic band structure, exchange interaction parameters, transport properties and p-d hybridization of the double layered  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  are significantly modified under on-site Coulomb correlation  $U$ , resulting in the observed magnetic ground state and transport anisotropy. The comparison with the band characteristics of the 3D perovskite  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  sets these similar doped manganites apart by means of different role of the Coulomb correlation on magnetism and transport properties.

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**Figure 1:** Calculated LSDA Mn-d projected densities of states (in states/eV-unit cell) of FM  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  (solid line) and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  (dashed line). The Fermi level is located at 0 eV. The minority spin contributions are shown as negative states.

**Figure 2:** The Mn-d projected densities of states (in states/eV-unit cell) of FM  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  (solid line) and AFM  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  (dashed line) ground states from LSDA+U calculations with  $U=7$  eV. The Fermi level is located at 0 eV. The minority spin contributions are shown as negative states.

**Figure 3:** The calculated angular electron density distribution in the energy window of the corresponding majority spin  $t_{2g}$  states for (a)  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ ,  $U=0$ ; (b)  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ ,  $U=7$ ; (c)  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ ,  $U=0$  eV and (d)  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ ,  $U=7$  eV. Only one  $\text{MnO}_6$  octahedron is shown (for the double layered manganite it belongs to the upper layer of the bilayer).

**Table 1:** Total energy differences,  $\Delta E_{TOT}$  in meV; the interlayer exchange interaction parameters,  $J_{dd}$  in meV; the ratio of the  $3z^2 - r^2$  and  $x^2 - y^2$  orbital contributions to the DOS at  $E_F$ ,  $N_{x^2-y^2}/N_{3z^2-r^2}$ ; the ratio of the  $3z^2 - r^2$  and  $x^2 - y^2$  band widths,  $W_{x^2-y^2}/W_{3z^2-r^2}$ ; the percentage Mn  $e_g$  and O- $p$  contributions to the DOS at  $E_F$ , N, for  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  and  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ .

	$\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$		$\text{LaSr}_2\text{Mn}_2\text{O}_7$	
	U=0	U=7	U=0	U=7
$\Delta E_{TOT}$	+338.9	+473.5	+91.0	-112.4
$J_{dd}$	+15.28	+94.50	+7.53	-1.84
$N_{x^2-y^2}/N_{3z^2-r^2}$	1.04	0.95	0.94	3.18
$W_{x^2-y^2}/W_{3z^2-r^2}$	1.00	1.00	1.24	2.38
$N(\text{Mn-}d_{3z^2-r^2})$	30	24	34	10
$N(\text{Mn-}d_{x^2-y^2})$	34	22	32	32
$N(\text{O1}_{apical}(\text{O2}_{apical})-p_z)$	12	18	2 (8)	0 (0)
$N(\text{O3}_{planar})-p_x/p_y)$	24	36	24	58





